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A SELF-HEALING POLYMER COMPOSITE FOR EXTENDED FATIGUE LIFE

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A novel approach is explored for improving the fatigue life of thermosetting polymers through the addition of self-healing functionality. Thermosetting polymers are used in a wide variety of applications, but are susceptible to the initiation and propagation of small cracks deep within the structure where detection is difficult and repair is virtually impossible. The material under investigation is an epoxy matrix composite, which utilizes embedded microcapsules to store a healing agent and an embedded catalyst. A propagating crack exposes particles of catalyst and ruptures the microcapsules, which release healing agent into the crack plane. Polymerization of the healing agent is triggered by contact with the catalyst. Fatigue crack retardation and arrest from self-healing functionality result from crack-tip shielding mechanisms, such as hydrodynamic pressure and artificial-crack closure. *In situ* healing is observed to significantly extended fatigue life or permanently arrested fatigue crack growth over a wide range of loading conditions.

A SELF-HEALING POLYMER COMPOSITE FOR EXTENDED FATIGUE LIFE

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Summary A self-healing polymer composite has been developed based on embedded microcapsules that deliver a chemical healing agent and catalyst. The autonomic response of this self-healing polymer to Mode-I fatigue loading is reported. Significant fatigue life extension and permanent fatigue crack arrest are obtained.

A novel approach is explored for improving the fatigue life of thermosetting polymers through the addition of self-healing functionality. Thermosetting polymers are used in a wide variety of applications ranging from composite structures to adhesive joints to microelectronic packaging. Due to their low strain-to-failure these polymers are highly susceptible to damage in the form of cracks. Fatigue loading is particularly problematic, giving rise to the initiation and propagation of small cracks deep within the structure where detection is difficult and repair is virtually impossible. These cracks often lead to catastrophic failure of the material. We utilize a strategy based on recent developments in self-healing technology [1-3] to autonomically repair fatigue cracks and extend the service-life of many polymeric components. The material under investigation is an epoxy matrix composite (EPON 828 cured with 12pph DETA), which utilizes embedded microcapsules [4] to store a healing agent and embedded Grubbs catalyst [5]. A propagating crack exposes particles of catalyst and ruptures the microcapsules, which release healing agent into the crack plane. Polymerization of the healing agent is triggered by contact with the catalyst, reestablishing structural integrity across the crack plane. The fatigue-crack propagation behavior is investigated using the tapered double-cantilever beam (TDCB) specimen with constant range of applied stress intensity factor ΔK. All measurements are made with a loading frequency of 5 Hz and load ratio $R = K_{min}/K_{max}$ of 0.1. Samples are cast in a silicon mold using a cure cycle of 24 hrs at room temperature followed by 24 hrs at 30 C and then precracked with a razor blade.

FATIGUE CRACK GROWTH PRECLUDING SELF-HEALING

Fatigue crack propagation in neat epoxy, epoxy with embedded microcapsules, and epoxy with embedded catalyst is accurately captured by the Paris power law. The effect of embedded microcapsule size and concentration on fatigue crack growth is shown in Fig. 1. The addition of microcapsules significantly reduces the crack growth rate above a transition ΔK_T. Above the transition, the Paris law exponent n is strongly dependent on the content of microcapsules, varying from 9.7 for neat epoxy to approximately 4.5 above 10 wt% microcapsules. Similar retardation behavior has been reported for epoxy with embedded rubber particles [6].

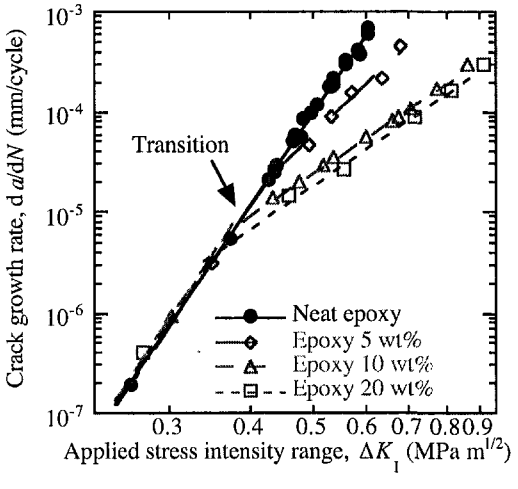

Figure 1. Influence of microcapsule concentration on the fatigue crack growth behavior for 180 μm diameter microcapsules.

IN SITU SELF-HEALING

Fatigue crack retardation and arrest from self-healing functionality result from crack-tip shielding mechanisms. Hydrodynamic pressure generated in flowing healing agent reduces both the loading and unloading of the crack tip. Moreover, the fully cured healing agent forms a polymer wedge at the crack tip, healing the crack as measured by negative crack growth, and preventing loading of the crack tip through adhesive mechanisms and unloading the crack tip through artificial-crack closure. Healing efficiency of the self-healing epoxy under cyclic loading is characterized with a fatigue-life-extension protocol [7],

$$\lambda = \frac{N_{\text{healed}} - N_{\text{control}}}{N_{\text{control}}}, \tag{1}$$

where N_{healed} is the total number of cycles to failure for the self-healing sample and N_{control} number of cycles to failure for a similar sample without healing. Under high-cycle fatigue (moderate ΔK_{I}), Fig. 2a, and low-cycle fatigue (high ΔK_{I}) when a rest period was employed, *in situ* healing extended fatigue life though temporary crack arrest and retardation. *In situ* self-healing permanently arrested crack growth under low-cycle fatigue conditions at low ΔK_{I}, Fig. 2b, and at moderate ΔK_{I} when a rest period was employed.

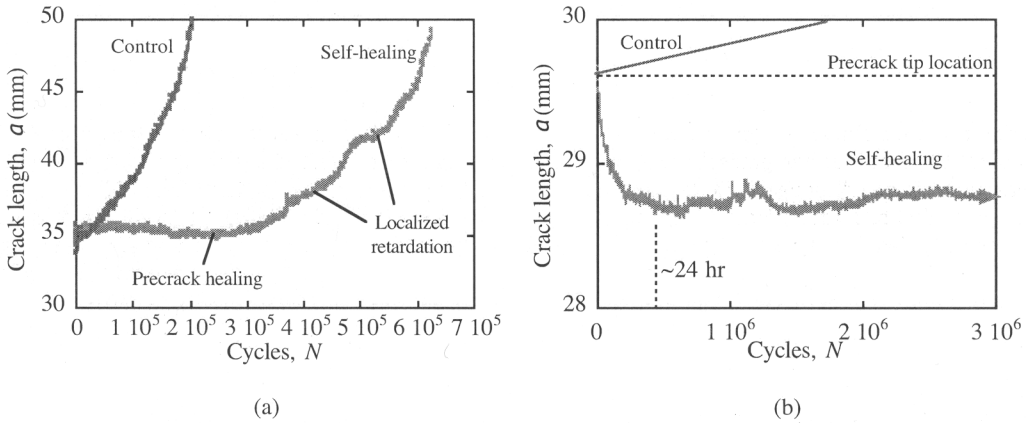

Figure 2. Crack length against fatigue cycles of *in situ* sample tested to failure in (a) the high-cycle fatigue regime, $\lambda = 213\%$. $\Delta K_{\text{I}} = 0.338$ MPa m$^{1/2}$ and (b) the threshold regime, $\lambda = \infty$. $\Delta K_{\text{I}} = 0.270$ MPa m$^{1/2}$.

CONCLUSIONS

A self-healing polymer is developed that improves the reliability of thermosetting polymers by initiating a healing process in response to damage. Fatigue-life extension is achieved by a combination of crack-tip shielding mechanisms induced by the self-healing functionality. First, viscous flow of the healing agent in the crack plane retards the crack growth. Second, as the healing agent fully polymerizes, a short-lived adhesive effect prevents loading of the crack and a long term crack closure effect preventing unloading of the crack tip reduces the crack length and retards additional crack growth. A material able to autonomically respond to fatigue crack growth represents a milestone in the development of safer, longer-lasting materials.

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